Program—Symposium F: Controlling the Interaction between Light and Semiconductor Nanostructures for Energy Applications

2014 MRS Spring Meeting & Exhibit

April 21-25, 2014
San Francisco, California

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2014-04-22

Symposium F

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- James F. Cahoon, University of North Carolina at Chapel Hill
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UNC Energy Frontier Research Center EFRC: Center for Solar Fuels at the University of North Carolina at Chapel Hill
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F1: Carrier Dynamics and Transport

- Chair: Charles Black
- Chair: Sorin Melinte
- Tuesday AM, April 22, 2014
- Marriott Marquis, Golden Gate Level, C

8:30 AM - *F1.01

Engineered Multi-Carrier Interactions in Semiconductor Nanocrystals for Light Emitting Diodes and Solar Cells

Victor I. Klimov1.

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9:00 AM - F1.02

Optimization of Bi-Exciton Binding Energy in CdSe/CdTe Core/Shell M.E.G. Solar Cells

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Hide Abstract

In a conventional solar cell the energy of an absorbed photon in excess of the bandgap is wasted as heat. Multiple exciton generation (MEG) in colloidal quantum dots (QDs) uses this energy to instead produce additional free charges, increasing the photocurrent and cell efficiency[1]. Theoretical predictions indicate that MEG has the potential to enhance the efficiency of a single gap cell from 33% to 42% [2], by minimisation of the energy threshold for MEG. An attractive interaction between excitons reduces the threshold by the biexciton binding energy, Bxx. This has been found to be small (-10meV) for type I QDs [3]. Previous calculations of Bxx in type II CdSe/CdTe QDs have found a large repulsion between excitons [4]. Here, we show that that a CdSe/CdTe core/shell QD exhibit large values of Bxx<0. Our theoretical methodology is based on an 14-band k.p Hamiltonian, with correct atomistic symmetry, C2v, of the zinc-blend material, which incorporates the effects of band mixing between the p-bonding, s-anti-bonding and p-anti-bonding states, SO interaction, crystal-field splitting, strain between core/shells and piezoelectric potentials [5]. Excitonic states were found using the full CI method, that incudes explicitly the effects of Coulomb interaction, exact exchange and correlations between many-electron configurations. Particular attention was paid to accurate modeling of the dielectric constant variation through the structure and surface polarization effects on core/shell and shell/solvent interfaces. Relevant material parameters are predicted using ab initio time-dependent density functional theory [6]. We conclude that: (i) it is not possible to predict biexciton binding using the Hartree approximation alone; it can only be predicted with a full CI Hamiltonian; (ii) CI predicts Bxx=-70 meV for QDs with 0.5 nm thick shell; (iii) by ignoring the dielectric confinement, it is not possible to predict biexciton binding for structures with shell thickness >0.75 nm; (iv) by changing the solvent dielectric constant from 1 to 2 the variation in the Bxx binding energy is as big as 100 meV; (v) a proper calculation of Bxx requires the inclusion of correlations and surface polarization effects but the effect of self-polarization is negligible. The strong biexciton binding found is explained by a stronger reduction in the Columbic repulsion between holes than reduction in the attraction between electrons and holes on the addition of the CdTe shell layer, which is a consequence of 4 fold degeneracy of the h-ground state imposed by symmetry of the structure.

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[2] V.I. Klimov, Appl Phys Lett 89, 123118 (2006)
[3]R. D. Schaller, et al, Nano Lett. 7, 3469 (2007)
[4] A. Piryatinski at al, Nano Lett 7, 108 (2007)
[5] S Tomic at al, J. Appl Phys 110, 053710 (2011)
[6] L. Bernasconi, S. Tomic et al, Phys Rev B 83, 195325 (2011)